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Princeton University SCHOOL OF ENGINEERING AND APPLIED SCIENCE
DEPARTMENT OF AEROSPACE AND MECHANICAL SCIENCES
GUGGENHEIM AEROSPACE PROPULSION LABORATORIES

May 15, 1969

Dr. Bernard T. Wolfson
Propulsion Division
Directorate of Engineering Sciences
Air Force Office of Scientific Research (SREP)
1400 Wilson Blvd.
Arlington, Virginia 22209

JUN 10 1969

Dear Dr. Wolfson:

The following comprises the final report on Contract AF 49(638)-1267 entitled, "Research on the Ignition of Solid Propellants." The subject contract was initiated on October 1, 1963 and terminated on September 30, 1968. Throughout this period the results of the work performed under this contract have been made available to the sponsor and to the technical community in general in the form of Princeton University AMS Reports and as articles in various journals; a bibliographic compilation of these publications is appended to this report. Since the details of the accomplishments of this program are thus available elsewhere, the present report is intended primarily as a summary of the knowledge gained during the contract period.

The present work is part of a continuing program designed to lead to understanding of the mechanism of solid propellant ignition, that is, to elucidate the sequence of physical and chemical processes comprising the ignition of a solid propellant. This program was initiated at Princeton in 1958 and has been supported by various AFOSR Grants and Contracts; subsequent to the termination of the subject contract, this program has been supported by AFOSR Grant No. 69-1651.

The specific areas of research to be reported on here fall under several headings, as follows: 1) ignition of solid fuels and solid propellants by heat conduction (shock tube); 2) theory of ignition in steady and non-steady diffusion flames; 3) ignition by intense radiation (experiments with CW IR laser); 4) theory of ignition by radiation; 5) ignition in a convective flow field (shock tunnel experiments). The general objective of the work in all of these areas is an increased understanding of ignition in a rocket motor environment which will yield ultimately, design principles and rules useful to the rocket motor engineer.

Continued on next page.....

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Ignition of Solid Fuels and Solid Propellants by Heat Conduction (Shock Tube):

One of the most important issues in the field of solid propellant ignition research is that of the ignition mechanism -- specifically, the identification and description of the reactions that control the process of thermal runaway. The use of a controllable and well-defined heat input to the propellant is a vital part of such research; if this requirement is not fulfilled, the results will be unintelligible. The shock tube, with a planar propellant sample mounted on the end wall of the driven section, provides a purely conductive heat input which fulfills this requirement. This tool has been used extensively throughout the present research program.

The initial shock tube data of McAlevy⁽¹⁾ strengthened the indications first obtained by other investigators^(2,3) that composite propellant ignition cannot be adequately described by a purely solid phase reaction model. His end wall ignition results demonstrated a strong dependence of ignition delay time on the amount of oxygen available in the adjacent test gas and on the total pressure of this gas. This work led to the hypothesis that the reaction sequence controlling thermal runaway occurs in the gas phase immediately above the propellant surface. Since these early results showed no enhancement of ignition by the presence of a condensed phase oxidizer such as ammonium perchlorate, it was concluded that these gaseous reactions involved pyrolyzed fuel vapor and the externally supplied oxygen only. These observations formed the basis for the original treatment of the gas phase ignition theory; the progress toward obtaining solutions to this model is described in more detail below under a separate heading.

Azcarraga⁽⁴⁾ obtained further experimental results consistent with the gas phase theory in a series of tests designed primarily to ascertain the effect of fuel volatility on ignition delay time. A group of aliphatic hydrocarbons with varied but generally high volatility showed little or no systematic dependence of ignition delay on volatility; however, fuels with appreciably lesser volatility did exhibit a strong correlation between this factor and ignition delay, the less volatile fuels being more difficult to ignite.

These results and those of Kurylko⁽⁵⁾ which showed no evidence of extensive participation of the solid oxidizer in the ignition event were encouraging in that they were consistent with the proposed gas phase ignition mechanism. However, when it came to the question of whether other reaction mechanisms^(6,7) (e.g., heterogeneous reactions as proposed by Anderson, et al,^(6,7)) could explain the same data, more precise data became necessary.

This question of reaction mechanism soon revolved around the precise way in which ignition depended on the amount of externally supplied oxygen. The theoretical work of Hermance^(8,9), to be discussed in the next section, showed that the gas phase theory predicted differing dependencies of ignition delay on oxygen partial pressure when this parameter was varied by changing the oxygen

percentage only and when it was varied by changing the total pressure only. Shannon⁽¹⁰⁾, however, obtained results which indicated the ignition delay depends only on oxygen partial pressure per se, regardless of how it is varied. This was consistent with previously published predictions of the heterogeneous theory. However, some question existed as to the applicability of these results and it was decided both to reproduce Shannon's experiments and further analyze the heterogeneous theory. After refinements in experimental techniques, Kashiwagi⁽¹¹⁾ showed that there are indeed significant but small differences in the ignition delay dependencies when oxygen partial pressure is varied in the two different ways mentioned above. At the same time Waldman⁽¹²⁾ showed that the heterogeneous theory as well as the gas phase theory predicted these differing dependencies. This work thus came to a disappointing end since it became evident that end-wall shock tube data could not distinguish between the two ignition mechanisms. It appears that the additional parameter provided by a hot flowing gas past the igniting sample may provide the necessary clues to resolve this question of mechanism. This work is proceeding via the use of a shock tunnel and will be discussed below.

Theory of Ignition in Steady and Non-Steady Diffusion Flames:

This work has been concerned primarily with the development and refinement of solutions to the gas phase ignition theory; more recently some work has also been done on diffusion flames in boundary layers.

Concurrent with their initial hypothesis that propellant ignition is controlled by a gas phase reaction mechanism, McAlevy and Summerfield⁽¹³⁾ developed a solution to their model based on several simplifying assumptions. The nature of the assumptions necessary to permit an analytical solution was such that the range of validity of this solution was rather severely limited. Nevertheless, the general qualitative consistency between theory and experiment was encouraging. In view of the complex, non-linear character of the full set of equations comprising the model, numerical solutions were obviously required to provide more general and exact predictions. The initial work of Hermance⁽⁸⁾ in this area was concerned with two restricted special cases: the first assumed that the solid fuel phase evaporated at a constant rate throughout the ignition interval; the second assumed that the fuel mass fraction at the solid/gas interface was fixed during this interval. Both of these assumptions decouple the events in the gas phase from the solid and eliminate the element of feedback by which the fuel vaporization rate is accelerated during ignition. Consequently these solutions are limited to fuels having relatively high volatility but they proved to be quite valuable in elucidating the dependence of ignition delay on environmental factors and the ignition criterion, as well. Later, Hermance⁽⁹⁾ obtained solutions to the full set of coupled equations including the effects of heat feedback to the solid fuel. These results showed the dramatic effect that this feedback has in shortening the ignition delay of nonvolatile fuels. They comprise the theoretical predictions that have been compared to the experimental shock tube data.

Numerical solutions of this type comprise the most precise solutions available to the complex set of differential equations describing gas phase ignition; however, they lack the generality and predictive capability of analytical solutions. Each change in one of the several relevant parameters requires a computer run. Analytical solutions, even for limited ranges of parameters, are thus being sought. Waldman⁽¹⁴⁾ carried out a perturbation analysis (for small time) of the gas phase ignition model subject to the assumption that the fuel surface temperature remains constant throughout the ignition interval (this decouples the gas from the solid). The solutions thus obtained are valid for highly volatile fuels and show excellent agreement (within this valid range) with the numerical solutions of Hermance. The more general problem involving a coupled solid-gas system is extremely difficult and remains for future work.

In conjunction with the experimental shock tube work of Kashiwagi, Waldman performed an analysis of the heterogeneous ignition theory as applied to this situation. By adapting a previous analysis by Williams⁽¹⁵⁾, he was able to show, as was described above, that the heterogeneous and gas phase theories yield qualitatively similar predictions for the shock tube ignition situation and thus cannot be distinguished by such tests.

There exists a large class of two-dimensional diffusion flame problems which are steady in the overall sense but unsteady from the viewpoint of any given reactant particle as it moves through the reaction field. Steady combustion in a laminar boundary layer is such a problem; it is mathematically similar to the gas phase ignition problem and is of importance in a variety of applications, such as hybrid rocket motors, re-entry vehicles, etc. Previous solutions to this type of problem were based on the assumption of infinitely fast reaction kinetics, which artificially collapses the combustion zone to a flame sheet. Waldman⁽¹⁴⁾ was able to relax this assumption somewhat for the diffusion flame in the laminar boundary layer over a wedge. Using the method of matched asymptotic expansions, he developed an expression for the flame thickness along the wedge resulting from non-infinite reaction rates and demonstrated the effects of this finite thickness on wedge burning rate and combustion efficiency. The general problem posed by the coupling between the chemistry, the flow, and the vaporization of the solid fuel is extremely complex and has been only partially solved. However, it is believed that the analysis performed will greatly facilitate future numerical and analytical solutions to this class of problems.

Ignition by Intense Radiation (Experiments with CW IR Laser):

The use of a purely radiative ignition stimulus both in propellant ignition mechanism research and in ignitability rating has become quite popular. This is so because such radiation sources as the arc image furnace provide a readily measured and controlled heat flux to the propellant surface. However, the interaction between this heat flux and the propellant can differ considerably from that produced by the predominantly convective heat flux usually

encountered in a rocket motor ignition situation. The present line of research was undertaken in an effort to elucidate the influence of the unique factors operant in radiative ignition.

The problem of analyzing the sequence of events comprising ignition of a composite propellant subjected to an essentially black-body radiation flux is an extremely formidable one. The usual complications posed by the complex physical and chemical nature of the propellant are compounded by the interaction between the polychromatic radiation beam and the optically heterogeneous propellant. Ohlemiller⁽¹⁶⁾ has shown that this radiation interaction problem can be broken into several classes in accord with the relative values of three characteristic lengths: the oxidizer particle size, the thickness of the thermal conduction wave, and the radiation penetration depth in the continuous fuel phase of the propellant. Most propellants fall into the class where all three of these lengths are comparable. This class poses a combined three-dimensional scattering and heat conduction problem for which only rough approximate solutions are possible.

It was decided that our theoretical concepts regarding some of the unique factors in radiative ignition would be best tested first in a simpler and better defined situation. The development of the high power continuous wave CO₂ laser provided a radiation source superior in several respects to the arc image furnace; one of its major advantages is the monochromatic character of the radiation beam which greatly simplifies the process of accounting for the fate of the radiation. A major problem with such a source, however, is the non-uniform flux distribution which it provides. An optical system was developed which provides a greatly improved, though not perfect, uniformity.

The initial work with this laser ignition apparatus has been confined to a study of the ignition of pure fuels in an oxidizing atmosphere. Ohlemiller examined the effect of pressure, radiation flux, percent oxygen in the test gas, and the radiation absorption coefficient of the fuel on the ignition behavior of two polymeric fuels. The results were found to be generally consistent with a gas phase ignition mechanism and in agreement with the theoretical predictions discussed in the next section. This work demonstrated the considerable ambiguity inherent in the concept of an ignition temperature and the inadequacy of applying low heating rate polymer pyrolysis data to the highly transient ignition situation. It demonstrated also the inapplicability of any other simple concept, such as ignition energy. Present work is being directed toward extending these results to the more complex situation posed by propellant ignition as against pure fuel ignition.

Theory of Ignition by Radiation:

Detailed mathematical modelling of composite propellant ignition by a monochromatic or polychromatic radiation beam is, of course, possible in principle. However, in view of the present limited state of knowledge of propellant physical and chemical parameters, such modelling is of limited value; the number of unknown parameters in such models is large enough to permit a forced

fit to a large body of experimental data, but such a procedure does not add much to the credibility of the model. A more modest approach of identifying and semi-quantitatively describing some of the major factors that influence radiative ignition behavior has been adopted in our initial theoretical work.

Probably the two most influential factors unique to radiative ignition are the presence of a cold gas adjacent to the sample surface and the instantaneous in-depth heating by penetration of the radiation. (17) The first of these can be properly assessed only within the context of a detailed ignition model because it is the chemical reaction rates themselves which are influenced. The second is more amenable to treatment, even though, as was mentioned in the previous section, radiation penetration in a composite propellant is too complex to permit precise description at the present time. By considering the progressively asymptotic cases in which chemical and diffusion processes, in-depth absorption, and heat conduction, respectively, dominate the radiative ignition delay, Summerfield and Ohlemiller (17) obtained an approximate expression for this delay. This expression has been shown to provide a very good semi-empirical correlation of nearly all available arc image furnace data. In particular, since it contains an explicit term accounting for propellant transparency, it permits one to deduce a correction (admittedly only approximate) for arc furnace data for this factor, which could otherwise cause considerable error in applying such data to a rocket motor ignition situation. This work (18) has now been incorporated into the ICRFG arc image ignition manual.

As derived, the approximate solution should best describe the radiative ignition of an optically homogeneous material, such as double-base propellant. Ohlemiller (16) tested this solution against his experimental laser-induced ignition data for pure fuels. The results showed that the solution provides a very good prediction of the effects of radiant flux and fuel absorptivity. With this basis, future work can now proceed on a more quantitative description of the fate of radiation incident on a composite propellant, incorporating the effects of both scattering and absorption in depth.

Ignition in a Convective Flow Field (Shock Tunnel Experiments):

As indicated above, it ultimately became evident that end wall shock tube ignition tests, in which a stagnant hot gas provides the ignition stimulus, could not provide the necessary information to distinguish between a heterogeneous and gas phase ignition mechanism. There is good reason to believe that the effects of a convective cross flow on both the length and position of the ignition delay can provide the necessary clues. Convective ignition is of further interest because it most closely models the rocket motor ignition situation.

Earlier work by various investigators has yielded rather confusing and inconsistent results. Bastress (19) and Hermance (8) found pronounced effects of sweep velocity on ignition delay; Ryan and Baer (20,21), however, found such effects only under

certain limited conditions. The present experiments are designed to produce a more precisely controlled boundary layer and to search more extensively for the dependence of ignition position on the various flow parameters. In addition, different boundary layer configurations, such as stagnation points, wakes, etc., will be examined.

A shock tunnel (4" I.D., about 70" long) has been designed and constructed for the above purpose and is in operation. The tunnel operates with O_2 and N_2 mixtures as the driven gas and He as the driver gas. The sweep² flow Mach number ranges typically from 0.1 to 1 with estimated experimental testing time of about 15 msec. The propellant sample is 2.5" long and 1.0" wide. A schlieren photography system and high speed cameras are used to obtain ignition behavior in the boundary layer. There are at present six photodiode detectors along the length of the propellant sample to determine where ignition occurs. Systematic testing is now in progress to determine at first the precise character of the boundary layer and heat transfer distribution over the sample surface as a function of pressure and flow velocity.

Supplementary Information:

Research will continue in several of these areas during the 1968-69 contract year under AFOSR Grant No. 69-1651. Unfortunately we will be unable to accomplish all of the objectives set forth in last year's Proposal because the level of funding has been reduced by about 40%. The main areas of work will be concerned with convective and radiative ignition. Systematic testing with the shock tunnel apparatus will be conducted to ascertain the detailed effects of heat flux, pressure, flow velocity, and oxygen concentration on the time and position of ignition for both pure fuels and propellants. This work, in conjunction with theoretical analysis of convective ignition, is expected to yield the necessary information needed to decide upon the controlling mechanism of ignition. The radiative ignition work will be concerned largely with carrying the knowledge gained in pure fuel ignition studies over into the analysis of propellant ignition by radiation. Efforts will be directed toward a quantitative description of the influences of scattering and in-depth absorption on ignition delay and the correction of experimental results for these factors by theoretical analysis.

Again, it should be noted that a much more detailed description of the work performed and the results achieved under the subject contract can be found in the individual reports published throughout the period of this contract as the work matured. The main publications were cited as references in the preceding text. A compendium of the reports and articles published under this and preceding contracts is presented in the Appendix, for convenience.

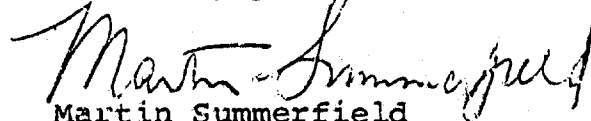
Three graduate students and one undergraduate student were supported in part by this program during the 1967-68 contract year. The first graduate student, Mr. T. Kashiwagi, has been actively engaged both in work on end-wall shock tube ignition and in the design and testing of the shock tunnel ignition apparatus. The

second graduate student, Mr. T. J. Ohlemiller, has been concerned with the experimental and theoretical analysis of radiative ignition. The third graduate student, Mr. C. H. Waldman, has been concerned with the theoretical analysis of steady and non-steady diffusion flames. The undergraduate student, Mr. R. B. Rothman, has been working in cooperation with Mr. Kashiwagi on the end-wall shock tube ignition studies.

Interest in the research work reported here has been expressed by various other groups. In particular, the ICRPG Arc Image Ignition Sub-Committee has shown continued interest in the analysis performed on radiative ignition. As was pointed out, much of this analysis has been incorporated into the arc image ignition handbook being compiled under the direction of Capt. C. E. Payne of AFRPL.

This completes our final report on Contract No. AF 49(638)-1267 entitled "Research on the Ignition of Solid Propellants."

Sincerely yours,



Martin Summerfield
Professor of Aerospace Propulsion

MS/eb

Enclosures: Appendix I
List of References

Appendix II
List of Reports

cc: Mr. P. L. Stang
Mr. S. Kidd

APPENDIX I

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9. ABSTRACT

The specific areas of research to be reported on here fall under several headings; as follows: 1) ignition of solid fuels and solid propellants by heat conduction (shock tube); 2) theory of ignition in steady and non-steady diffusion flames; 3) ignition by intense radiation (experiments with CW IR laser); 4) theory of ignition by radiation; 5) ignition in a convective flow field (shock tunnel experiments). The general objective of the work in all of these areas is an increased understanding of ignition in a rocket motor environment which will yield ultimately, design principles and rules useful to the rocket motor engineer.

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KEY WORDS

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